

FORM PTO-1390  
(REV 11-98)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371

Beiersdorf 665-KGB

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

09/700103

INTERNATIONAL APPLICATION NO.

PCT/EP99/02709

INTERNATIONAL FILING DATE

22. April 1999 (22.04.99)

PRIORITY DATE CLAIMED

9. Mai 1998 (09.05.98)

TITLE OF INVENTION

SEE APPENDIX

APPLICANT(S) FOR DO/EO/US

Heinrich GERS-BARLAG and Rainer KROPKE

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☒ has been transmitted by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
  - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☐ have been transmitted by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☐ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A **FIRST** preliminary amendment.  
☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☒ Other items or information:  
-International Search Report (one page)

17. ☐ The following fees are submitted:

BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):

Neither international preliminary examination fee (37 CFR 1.482)  
nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO  
and International Search Report not prepared by the EPO or JPO ..... \$970.00

International preliminary examination fee (37 CFR 1.482) not paid to  
USPTO but International Search Report prepared by the EPO or JPO ..... \$860.00

International preliminary examination fee (37 CFR 1.482) not paid to USPTO but  
international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... \$760.00

International preliminary examination fee paid to USPTO (37 CFR 1.482)  
but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... \$670.00

International preliminary examination fee paid to USPTO (37 CFR 1.482)  
and all claims satisfied provisions of PCT Article 33(1)-(4) ..... \$96.00

ENTER APPROPRIATE BASIC FEE AMOUNT =

CALCULATIONS PTO USE ONLY

\$ 860.00

Surcharge of \$130.00 for furnishing the oath or declaration later than ☐ 20 ☐ 30  
months from the earliest claimed priority date (37 CFR 1.492(e)).

\$

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE
Total claims	6 - 20 =	0	X \$18.00
Independent claims	2 - 3 =	0	X \$80.00
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$ 270.00

\$ 0

\$ 0

\$

TOTAL OF ABOVE CALCULATIONS =

\$ 860.00

Reduction of 1/2 for filing by small entity, if applicable. A Small Entity Statement  
must also be filed (Note 37 CFR 1.9, 1.27, 1.28).

\$

SUBTOTAL =

\$ 860.00

Processing fee of \$130.00 for furnishing the English translation later than ☐ 20 ☐ 30  
months from the earliest claimed priority date (37 CFR 1.492(f)).

\$

+

TOTAL NATIONAL FEE =

\$ 860.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be  
accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property

\$

+

0

TOTAL FEES ENCLOSED =

\$ 0

Amount to be:  
refunded

\$

charged

\$860.00

a. ☐ A check in the amount of \$\_\_\_\_\_ to cover the above fees is enclosed.

b. ☒ Please charge my Deposit Account No. 14-1263 in the amount of \$ 860.00 to cover the above fees.  
A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any  
overpayment to Deposit Account No. 14-1263. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR  
1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

Norris McLaughlin & Marcus PA  
220 East 42nd Street  
30th Floor  
New York, New York 10017  
(212) 808-0700

SIGNATURE:

Kurt G. Briscoe

NAME

33,141

REGISTRATION NUMBER

Beiersdorf 665-KGB  
6713-Dr. Wi-ka

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPLICANTS : HEINRICH GERS-BARLAG ET AL.  
SERIAL NO. : TO BE ASSIGNED  
FILED : HEREWITH  
FOR : COSMETIC AND DERMATOLOGICAL LIGHT PROTECTION  
FORMULATIONS CONTAINING TRIAZINE DERIVATIVES  
AND ONE OR MORE ESTERS OF UNBRANCHED-CHAIN  
CARBOXYLIC ACIDS AND BRANCHED-CHAIN ALCOHOLS  
ART UNIT : UNASSIGNED  
EXAMINER : UNASSIGNED

---

November 9, 2000

Hon. Commissioner of Patents  
Washington, D.C. 20231

**PRELIMINARY AMENDMENT**

SIR:

Prior to examination, please amend the above-identified application as follows:

IN THE SPECIFICATION:

Insert as the first sentence: -- This application is a 371 of PCT/EP99/02709 filed on

April 22, 1999. --

09/700103-1000

IN THE CLAIMS:

Claim 2, line 1, delete "The use of" and substitute -- Method of using --; and  
last line, delete "for the use".

Claims 3-6, line 1 in each, after "1" insert a comma, and delete the balance of the line.

REMARKS

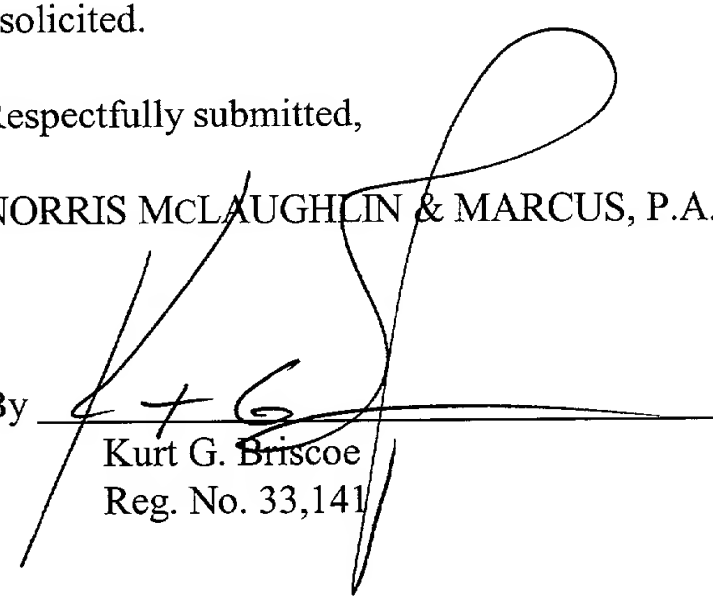
The amendments above eliminate multiple dependency, and convert the subject matter of  
claim 2 to a method of use.

Early and favorable action is earnestly solicited.

Respectfully submitted,

NORRIS MCLAUGHLIN & MARCUS, P.A.

By

  
Kurt G. Briscoe  
Reg. No. 33,141

220 East 42<sup>nd</sup> Street  
30<sup>th</sup> Floor  
New York, New York 10017  
(212) 808-0700

**Patent claims:** as submitted November 9, 2000

1. An active ingredient combination comprising tris(2-ethylhexyl) 4,4',4''-(1,3,5-triazine-2,4,6-triyltriimino)trisbenzoate and one or more esters of unbranched-chain carboxylic acids and branched-chain alcohols.
2. Method of using one or more esters of unbranched-chain carboxylic acids and branched-chain alcohols as solvent, solubility promoter or solubilizer for tris(2-ethylhexyl) 4,4',4''-(1,3,5-triazine-2,4,6-triyltriimino)trisbenzoate, in particular for the use in light protection compositions.
3. The combination as claimed in claim 1 , characterized in that the total amount of tris(2-ethylhexyl) 4,4',4''-(1,3,5-triazine-2,4,6-triyltriimino)trisbenzoate in the finished cosmetic or dermatological preparations is chosen from the range 0.1-10.0% by weight, preferably 0.5-6.0% by weight, in each case based on the total weight of the preparations.
4. The combination as claimed in claim 1 , characterized in that the ester(s) of branched-chain carboxylic acids and branched-chain alcohols is / are present in a mixture with the parent alcohols R<sub>2</sub>-OH.
5. The combination as claimed in claim 1 , characterized in that the ester of branched-chain carboxylic acids and branched-chain alcohols chosen is hexyldecyl laurate.
6. The combination as claimed in claim 1. , characterized in that the total amount of one or more esters of unbranched-chain carboxylic acids and branched-chain alcohols in the finished cosmetic or dermatological preparations is chosen from the range 0.1-25.0% by weight, preferably 0.5-15.0% by weight, in each case based on the total weight of the preparations.

Beiersdorf Aktiengesellschaft  
Hamburg

## Description

### **Cosmetic and dermatological light protection formulations containing triazine derivatives and one or more esters of unbranched-chain carboxylic acids and branched-chain alcohols**

The present invention relates to cosmetic and dermatological light protection formulations, in particular skincare cosmetic and dermatological light protection preparations.

The harmful effect of the ultraviolet part of solar radiation on the skin is generally known. While rays with a wavelength of less than 290 nm (the so-called UVC region) are absorbed by the ozone layer in the earth's atmosphere, rays in the range between 290 nm and 320 nm, the so-called UVB range, cause erythema, simple sunburn or even burns of varying severity.

The maximum given for the erythema activity of sunlight is the narrower range around 308 nm.

Numerous compounds are known for protecting against UVB radiation; these are usually derivatives of 3-benzylidenecamphor, of 4-aminobenzoic acid, of cinnamic acid, of salicylic acid, of benzophenone and also of 2-phenylbenzimidazole.

It is also important to have available filters for the range between about 320 nm and about 400 nm, the so-called UVA region, since its rays can likewise cause damage. Thus, it has been found that UVA radiation leads to damage of the elastic and collagenous fibers of connective tissue, causing premature aging of the skin, and that it

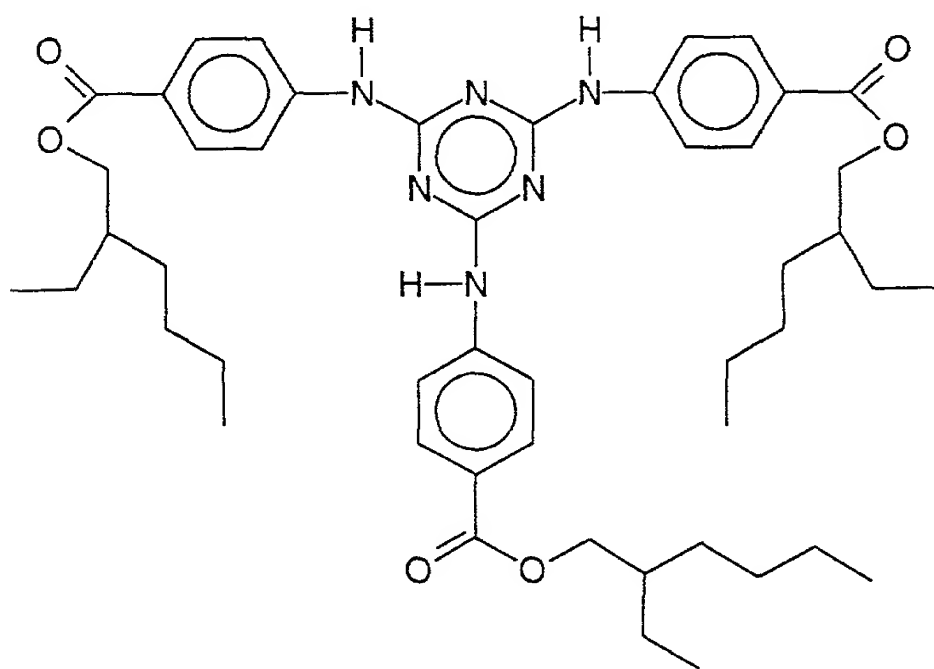
is to be regarded as a cause of numerous phototoxic and photoallergic reactions. The harmful effect of UVB radiation can be intensified by UVA radiation.

However, UV radiation can also lead to photochemical reactions, in which case the photochemical reaction products intervene in the skin's metabolism.

Such photochemical reaction products are predominantly free-radical compounds, e.g. hydroxyl radicals. Undefined free-radical photoproducts which form in the skin itself can also display uncontrolled secondary reactions because of their high reactivity. However, singlet oxygen, a non-radical excited state of the oxygen molecule, can also arise during UV radiation, as can short-lived epoxides and many other species. Singlet oxygen, for example, differs from the normal triplet oxygen (free-radical ground state) by virtue of its increased reactivity. However, excited reactive (free-radical) triplet states of the oxygen molecule also exist.

Furthermore, UV radiation is a type of ionizing radiation. There is therefore the risk that UV exposure may also produce ionic species which then, for their part, are capable of oxidative intervention in the biochemical processes.

An advantageous UVB filter is tris(2-ethylhexyl) 4,4',4''-(1,3,5-triazine-2,4,6-triyltriimino)trisbenzoate, also known as 2,4,6-tris-[anilino(p-carbo-2'-ethyl-1'-hexyloxy)]-1,3,5-triazine.

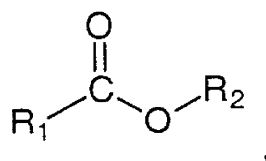


This UVB filter is sold by BASF Aktiengesellschaft under the trade name UVINUL7 T 150 and is notable for its good UV absorption properties.

The main disadvantage of this UVB filter is its poor solubility in lipids. Known solvents for this UVB filter can dissolve a maximum of about 15% by weight of this filter, corresponding to about 1-1.5% by weight of dissolved, and therefore active, UV filter substance.

It was nevertheless surprising, and could not have been foreseen by the person skilled in the art, that active ingredient combinations comprising tris(2-ethylhexyl) 4,4',4''-(1,3,5-triazine-2,4,6-triyltriimino)trisbenzoate and one or more esters of unbranched-chain carboxylic acids and branched-chain alcohols, or the use of one or more esters of unbranched-chain carboxylic acids and branched-chain alcohols as solvent, solubility promoter or solubilizer for tris(2-ethylhexyl) 4,4',4''-(1,3,5-triazine-2,4,6-triyltriimino)trisbenzoate, in particular for the use in light protection compositions, would overcome the disadvantages of the prior art.

The esters of unbranched-chain carboxylic acids and branched-chain alcohols according to the invention can advantageously be chosen from the group of substances of the general formula



in which  $\text{R}_1$  is an unbranched-chain alkyl radical having 3 to 30 carbon atoms, and  $\text{R}_2$  represents mono- or polybranched alkyl radicals having 3 to 30 carbon atoms.

$\text{R}_1$  is advantageously an unbranched-chain alkyl radical having up to 20 carbon atoms.

In addition,  $\text{R}_2$  is advantageously a mono- or polybranched alkyl radical having up to 25 carbon atoms.

For the purposes of the present invention, the carboxylic ester(s) is/are very particularly advantageously present in a mixture with the parent alcohols  $\text{R}_2\text{-OH}$ .



It is particularly advantageous to use hexyldecyl laurate, for example in a mixture with hexyldecyl alcohol as the carboxylic ester used according to the invention. Such a mixture is entered in the Chemical Abstracts under the registry numbers 70693-04-8 and 34362-27-1 and is sold, for example, by Henkel KGaA under the name Cetiol®PGL. As is known, it can be used in cosmetic and dermatological preparations as an oil component.

It was nevertheless surprising that the addition of one or more esters of unbranched-chain carboxylic acids and branched-chain alcohols effects stabilization of solutions of tris(2-ethylhexyl) 4,4',4''-(1,3,5-triazine-2,4,6-triyltriimino)trisbenzoate, since the latter substance not only has poor solubility, but also readily crystallizes out again from its solution. The invention therefore also provides a method of stabilizing solutions of tris(2-ethylhexyl) 4,4',4''-(1,3,5-triazine-2,4,6-triyltriimino)trisbenzoate, which comprises adding an effective content of one or more esters of unbranched-chain carboxylic acids and branched-chain alcohols to such solutions.

The total amount of tris(2-ethylhexyl) 4,4',4''-(1,3,5-triazine-2,4,6-triyltriimino)trisbenzoate in the finished cosmetic or dermatological preparations is advantageously chosen from the range 0.1-10.0% by weight, preferably 0.5-6.0% by weight, based on the total weight of the preparations.

The total amount of one or more esters of unbranched-chain carboxylic acids and branched-chain alcohols in the finished cosmetic or dermatological preparations is advantageously chosen from the range 0.1-25.0% by weight, preferably 0.5-15.0% by weight, based on the total weight of the preparations.

It is advantageous to choose weight ratios of tris(2-ethylhexyl) 4,4',4''-(1,3,5-triazine-2,4,6-triyltriimino)trisbenzoate and one or more esters of unbranched-chain carboxylic acids and branched-chain alcohols from the range 1 : 10 to 10 : 1, preferably 1 : 4 to 4 : 1.

Cosmetic and dermatological preparations according to the invention also advantageously comprise inorganic pigments based on metal oxides and/or other

For the purposes of the present invention, it is particularly advantageous, although not imperative, if the inorganic pigments are present in hydrophobic form, i.e. they have been surface-treated to repel water. This surface treatment can consist in providing the pigments with a thin hydrophobic layer by processes known per se.

$$n \text{ TiO}_2 + m (\text{RO})_3 \text{ Si-R}' \rightarrow n \text{ TiO}_2 (\text{surf.})$$

Advantageous TiO<sub>2</sub> pigments are available, for example, under the trade names MT 100 T from TAYCA, also M 160 from Kemira and T 805 from Degussa.

For use, the cosmetic and dermatological preparations according to the invention are applied to the skin and/or hair in sufficient amount and in the manner conventional for cosmetics.

Particularly preferred cosmetic and dermatological preparations are those which are in the form of a sunscreen. Advantageously, these can additionally comprise at least one

further UVA filter and/or at least one further UVB filter and/or at least one inorganic pigment, preferably an inorganic micropigment.

The cosmetic and dermatological preparations according to the invention can comprise cosmetic auxiliaries such as those customarily used in such preparations, e.g. preservatives, bactericides, perfumes, antifoams, dyes, pigments which have a coloring effect, thickeners, moisturizers and/or humectants, fats, oils, waxes, or other customary constituents of a cosmetic or dermatological formulation, such as alcohols, polyols, polymers, foam stabilizers, electrolytes, organic solvents or silicone derivatives.

An additional content of antioxidants is generally preferred. According to the invention, favorable antioxidants which can be used are all antioxidants suitable or customary for cosmetic and/or dermatological applications.

The antioxidants are advantageously chosen from the group consisting of amino acids (e.g. glycine, histidine, tyrosine, tryptophan) and derivatives thereof, imidazoles, (e.g. urocanic acid) and derivatives thereof, peptides, such as D,L-carnosine, D-carnosine, L-carnosine and derivatives thereof (e.g. anserine), carotenoids, carotenes (e.g.  $\alpha$ -carotene,  $\beta$ -carotene, lycopene) and derivatives thereof, chlorogenic acid and derivatives thereof, lipoic acid and derivatives thereof (e.g. dihydrolipoic acid), aurothioglucose, propylthiouracil and other thiols (e.g. thioredoxin, glutathione, cysteine, cystine, cystamine and glycosyl, N-acetyl, methyl, ethyl, propyl, amyl, butyl and lauryl, palmitoyl, oleyl,  $\gamma$ -linoleyl, cholesteryl and glyceryl esters thereof) and salts thereof, dilauryl thiodipropionate, distearyl thiodipropionate, thiodipropionic acid and derivatives thereof (esters, ethers, peptides, lipids, nucleotides, nucleosides and salts) and sulfoximine compounds (e.g. buthionine-sulfoximines, homocysteine-sulfoximine, buthionine-sulfones, penta-, hexa-, heptathionine-sulfoximines) in very low tolerated doses (e.g. pmol to  $\mu$ mol/kg), and also (metal) chelating agents (e.g.  $\alpha$ -hydroxyfatty acids, palmitic acid, phytic acid, lactoferrin),  $\alpha$ -hydroxy acids (e.g. citric acid, lactic acid, malic acid), humic acid, bile acid, bile extracts, bilirubin, biliverdin, EDTA, EGTA and derivatives thereof, unsaturated fatty acids and derivatives thereof (e.g.  $\gamma$ -linolenic acid, linoleic acid, oleic acid), folic acid and derivatives thereof, furfurylidenesorbitol and

derivatives thereof, ubiquinone and ubiquinol and derivatives thereof, vitamin C and derivatives (e.g. ascorbyl palmitate, Mg ascorbyl phosphate, ascorbyl acetate), tocopherols and derivatives (e.g. vitamin E acetate), vitamin A and derivatives (vitamin A palmitate) and coniferyl benzoate of benzoin resin, rutinic acid and derivatives thereof,  $\alpha$ -glycosylrutin, ferulic acid, furfurylidene-glucitol, carnosine, butylhydroxytoluene, butylhydroxyanisole, nordihydroguaiacic acid, nordihydroguaiaretic acid, trihydroxybutyrophenone, uric acid and derivatives thereof, mannose and derivatives thereof, zinc and derivatives thereof (e.g. ZnO, ZnSO<sub>4</sub>), selenium and derivatives thereof (e.g. selenomethionine), stilbenes and derivatives thereof (e.g. stilbene oxide, trans-stilbene oxide), and the derivatives (salts, esters, ethers, sugars, nucleotides, nucleosides, peptides and lipids) of said active ingredients which are suitable according to the invention.

The amount of the abovementioned antioxidants (one or more compounds) in the preparations is preferably from 0.001 to 30% by weight, particularly preferably from 0.05 to 20% by weight, in particular from 1 to 10% by weight, based on the total weight of the preparation.

If vitamin E and/or derivatives thereof are used as the antioxidant or antioxidants, their respective concentrations are advantageously chosen from the range 0.001 - 10% by weight, based on the total weight of the formulation.

If vitamin A or vitamin A derivatives or carotenes or derivatives thereof are used as the antioxidant or antioxidants, their respective concentrations are advantageously chosen from the range 0.001 - 10% by weight, based on the total weight of the formulation.

The lipid phase can advantageously be chosen from the following group of substances:

- mineral oils, mineral waxes
- oils, such as triglycerides of capric or of caprylic acid, but preferably castor oil;
- fats, waxes and other natural and synthetic fatty substances, preferably esters of fatty acids with alcohols of low carbon number, e.g. with isopropanol,

propylene glycol or glycerol, or esters of fatty alcohols with alkanolic acids of low carbon number or with fatty acids;

- alkyl benzoates;
- silicone oils such as dimethylpolysiloxanes, diethylpolysiloxanes, diphenylpolysiloxanes and mixtures thereof.

For the purposes of the present invention, the oil phase of the emulsions, oleogels and hydrodispersions or lipodispersions is advantageously chosen from the group of esters of saturated and/or unsaturated, branched and/or unbranched alkanecarboxylic acids having a chain length of from 3 to 30 carbon atoms and saturated and/or unsaturated, branched and/or unbranched alcohols having a chain length of from 3 to 30 carbon atoms, from the group of esters of aromatic carboxylic acids and saturated and/or unsaturated, branched and/or unbranched alcohols having a chain length of from 3 to 30 carbon atoms. Such ester oils can advantageously be chosen from the group consisting of isopropyl myristate, isopropyl palmitate, isopropyl stearate, isopropyl oleate, n-butyl stearate, n-hexyl laurate, n-decyl oleate, isooctyl stearate, isononyl stearate, isononyl isononanoate, 2-ethylhexyl palmitate, 2-ethylhexyl laurate, 2-hexyldecyl stearate, 2-octyldodecyl palmitate, oleyl oleate, oleyl erucate, erucyl oleate, erucyl erucate and synthetic, semi-synthetic and natural mixtures of such esters, e.g. jojoba oil.

The oil phase can also advantageously be chosen from the group of branched and unbranched hydrocarbons and hydrocarbon waxes, of silicone oils, of dialkyl ethers, from the group of saturated or unsaturated, branched or unbranched alcohols, and also fatty acid triglycerides, namely the triglycerol esters of saturated and/or unsaturated, branched and/or unbranched alkanecarboxylic acids having a chain length of from 8 to 24, in particular 12 - 18, carbon atoms. The fatty acid triglycerides can advantageously be chosen, for example, from the group of synthetic, semi-synthetic and natural oils, e.g. olive oil, sunflower oil, soybean oil, groundnut oil, rapeseed oil, almond oil, palm oil, coconut oil, palm kernel oil and the like.

For the purposes of the present invention, any mixtures of such oil and wax components can also advantageously be used. When required, it can also be

advantageous to use waxes, for example cetyl palmitate, as the sole lipid component of the oil phase.

The oil phase is advantageously chosen from the group consisting of 2-ethylhexyl isostearate, octyldodecanol, isotridecyl isononanoate, isoeicosane, 2-ethylhexyl cocoate, C<sub>12</sub>-C<sub>15</sub>-alkyl benzoate, caprylic/capric triglyceride and dicaprylyl ether.

Mixtures of C<sub>12</sub>-C<sub>15</sub>-alkyl benzoate and 2-ethylhexyl isostearate, mixtures of C<sub>12</sub>-C<sub>15</sub>-alkyl benzoate and isotridecyl isononanoate and mixtures of C<sub>12</sub>-C<sub>15</sub>-alkyl benzoate, 2-ethylhexyl isostearate and isotridecyl isononanoate are particularly advantageous.

For the purposes of the present invention, of the hydrocarbons, paraffin oil, squalane and squalene can advantageously be used.

The oil phase can advantageously also contain cyclic or linear silicone oils or can consist entirely of such oils, although it is preferable to use an additional content of other oil phase components in addition to the silicone oil or silicone oils.

Cyclomethicone (octamethylcyclotetrasiloxane) is advantageously used as silicone oil to be used according to the invention. However, other silicone oils can also be used advantageously for the purposes of the present invention, for example hexamethylcyclotrisiloxane, polydimethylsiloxane, poly(methylphenylsiloxane).

Mixtures of cyclomethicone and isotridecyl isononanoate and mixtures of cyclomethicone and 2-ethylhexyl isostearate are particularly advantageous.

The aqueous phase of the preparations according to the invention may advantageously comprise

- alcohols, diols or polyols of low carbon number, and also ethers thereof, preferably ethanol, isopropanol, propylene glycol, glycerol, ethylene glycol, ethylene glycol monoethyl or monobutyl ether, propylene glycol monomethyl, monoethyl or monobutyl ether, diethylene glycol monomethyl or monoethyl ether and analogous products, and also alcohols of low carbon number, e.g.

ethanol, isopropanol, 1,2-propanediol, glycerol, and in particular one or more thickeners which can advantageously be chosen from the group consisting of silicon dioxide, aluminum silicates, polysaccharides and derivatives thereof, e.g. hyaluronic acid, xanthan gum, hydroxypropylmethylcellulose, and particularly advantageously from the group of polyacrylates, preferably a polyacrylate from the group of the so-called Carbopols, for example Carbopol grades 980, 981, 1382, 2984, 5984, in each case individually or in combination.

The cosmetic or dermatological light protection preparations advantageously comprise inorganic pigments, in particular micropigments, e.g. in amounts of from 0.1% by weight to 30% by weight, preferably in amounts of from 0.5% by weight to 10% by weight, but in particular from 1% by weight to 6% by weight, based on the total weight of the preparations.

It is advantageous according to the invention to use, apart from the combinations according to the invention, oil-soluble UVA filters and/or UVB filters in the lipid phase and/or water-soluble UVA filters and/or UVB filters in the aqueous phase.

The light protection formulations according to the invention can advantageously comprise further substances which absorb UV radiation in the UVB region, the total amount of filter substances being e.g. from 0.1% by weight to 30% by weight, preferably from 0.5 to 10% by weight, in particular from 1 to 6% by weight, based on the total weight of the preparations, in order to provide cosmetic preparations which protect the skin from the entire range of ultraviolet radiation. They can also be used as sunscreens.

The further UVB filters can be oil-soluble or water-soluble. Examples of advantageous oil-soluble UVB filter substances are:

- 3-benzylidenecamphor derivatives, preferably 3-(4-methylbenzylidene)camphor, 3-benzylidenecamphor;
- 4-aminobenzoic acid derivatives, preferably 2-ethylhexyl 4-(dimethylamino)benzoate, amyl 4-(dimethylamino)benzoate;
- esters of cinnamic acid, preferably 2-ethylhexyl 4-methoxycinnamate, isopentyl 4-methoxycinnamate;

- derivatives of benzophenone, preferably 2-hydroxy-4-methoxybenzophenone, 2-hydroxy-4-methoxy-4'-methylbenzophenone, 2,2'-dihydroxy-4-methoxybenzophenone;
- esters of benzalmalonic acid, preferably di(2-ethylhexyl) 4-methoxybenzalmalonate.

Examples of advantageous water-soluble UVB filter substances are:

- salts of 2-phenylbenzimidazole-5-sulfonic acid, such as its sodium, potassium or its triethanolammonium salt, and also the sulfonic acid itself;
- sulfonic acid derivatives of benzophenones, preferably 2-hydroxy-4-methoxybenzophenone-5-sulfonic acid and salts thereof;
- sulfonic acid derivatives of 3-benzylidenecamphor, such as e.g. 4-(2-oxo-3-bornylidenemethyl)benzenesulfonic acid, 2-methyl-5-(2-oxo-3-bornylidenemethyl)sulfonic acid and salts thereof.

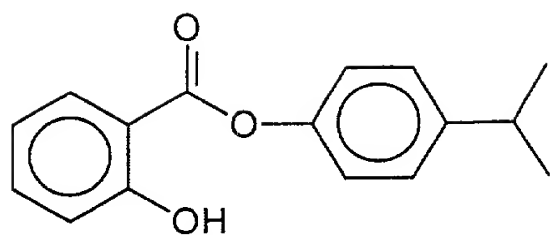
The list of said further UVB filters which can be used in combination with the active ingredient combinations according to the invention is of course not intended to be limiting.

It can also be advantageous to combine the combinations according to the invention with further UVA filters which have hitherto been customarily present in cosmetic preparations. These substances are preferably derivatives of dibenzoylmethane, in particular 1-(4'-tert-butylphenyl)-3-(4'-methoxyphenyl)propane-1,3-dione and 1-phenyl-3-(4'-isopropylphenyl)propane-1,3-dione. These combinations and preparations which contain these combinations are also provided by the invention. The amounts which may be used are as for the UVB combination.

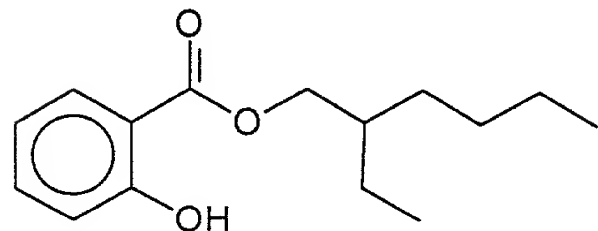
It is further advantageous to combine the active ingredient combinations according to the invention with further UVA and/or UVB filters.

It is also particularly advantageous to combine the active ingredient combinations according to the invention with salicylic acid derivatives, some of which, as is known, can also absorb UV radiation. Customary UV filters include

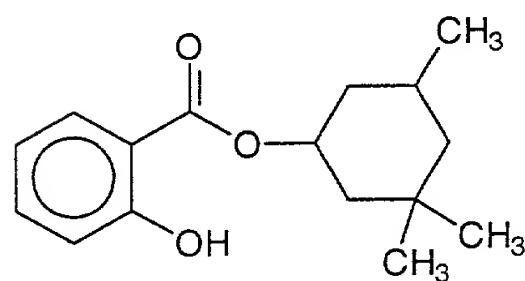




(4-isopropylbenzyl salicylate),



(2-ethylhexyl salicylate, octyl salicylate),



(homomenthyl salicylate).

The invention further provides a process for the preparation of the cosmetic and/or dermatological light protection preparations according to the invention, which comprises suspending and, if desired, homogenizing tris(2-ethylhexyl) 4,4',4''-(1,3,5-triazine-2,4,6-triyltriimino)trisbenzoate in one or more esters of unbranched-chain carboxylic acids and branched-chain alcohols or an oil phase containing one or more esters of unbranched-chain carboxylic acids and branched-chain alcohols with uniform stirring and if necessary with warming, where appropriate combining the mixture with further lipid components and optionally with one or more emulsifiers, then mixing the oil phase with the aqueous phase into which a thickener has optionally been incorporated and which is preferably at the same temperature as the oil phase, if desired homogenizing the mixture, and allowing it to cool to room temperature. After the mixture has cooled to room temperature, homogenization may be repeated, particularly if volatile constituents are still to be incorporated.

The examples below serve to illustrate the present invention without limiting it. Unless indicated otherwise, all amounts, proportions and percentages are

based on the weight and the total amount or on the total weight of the preparations.

### **Example 1**

O/W emulsion

	% by weight
Stearic acid	3.50
Glycerol	3.00
Cetylstearyl alcohol	0.50
Dicaprylyl ether	8.00
Uvinul®T150	5.00
Hexyldecyl laurate	12.00
Sodium hydroxide (45% strength)	0.33
Carbomer	0.20
Preservative	q.s.
Perfume	q.s.
Water, demin.	ad 100.00

### **Example 2**

W/O emulsion

	% by weight
Arlacel®989	5.50
Butylene glycol	5.00
Hexyldecyl laurate	12.00
Uvinul®T150	5.00
Cetylstearyl isononanoate	6.00
Carbomer	0.20
Preservative	q.s.
Perfume	q.s.
Water, demin.	ad 100.00

## Hydrodispersion gel

	% by weight
Carbomer	0.50
Butylene glycol	5.00
Hexyldecyl laurate	10.00
Sodium hydroxide (45% strength)	0.35
Uvinul®T150	5.00
Hydroxypropylcellulose	0.60
Preservative	q.s.
Perfume	q.s.
Water, demin.	ad 100.00

**Patent claims:**

1. An active ingredient combination comprising tris(2-ethylhexyl) 4,4',4''-(1,3,5-triazine-2,4,6-triyltriimino)trisbenzoate and one or more esters of unbranched-chain carboxylic acids and branched-chain alcohols.
2. The use of one or more esters of unbranched-chain carboxylic acids and branched-chain alcohols as solvent, solubility promoter or solubilizer for tris(2-ethylhexyl) 4,4',4''-(1,3,5-triazine-2,4,6-triyltriimino)trisbenzoate, in particular for the use in light protection compositions.
3. The combination as claimed in claim 1 or the use as claimed in claim 2, characterized in that the total amount of tris(2-ethylhexyl) 4,4',4''-(1,3,5-triazine-2,4,6-triyltriimino)trisbenzoate in the finished cosmetic or dermatological preparations is chosen from the range 0.1-10.0% by weight, preferably 0.5-6.0% by weight, in each case based on the total weight of the preparations.
4. The combination as claimed in claim 1 or the use as claimed in claim 2, characterized in that the ester(s) of branched-chain carboxylic acids and branched-chain alcohols is / are present in a mixture with the parent alcohols  $R_2-OH$ .
5. The combination as claimed in claim 1 or the use as claimed in claim 2, characterized in that the ester of branched-chain carboxylic acids and branched-chain alcohols chosen is hexyldecyl laurate.
6. The combination as claimed in claim 1 or the use as claimed in claim 2, characterized in that the total amount of one or more esters of unbranched-chain carboxylic acids and branched-chain alcohols in the finished cosmetic or dermatological preparations is chosen from the range 0.1-25.0% by weight, preferably 0.5-15.0% by weight, in each case based on the total weight of the preparations.

**Abstract:**

Active ingredient combinations comprising tris(2-ethylhexyl) 4,4',4''-(1,3,5-triazine-2,4,6-triyltriimino)trisbenzoate and one or more esters of unbranched-chain carboxylic acids and branched-chain alcohols.

## COMBINATION DECLARATION & POWER OF ATTORNEY

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled „**Cosmetic and dermatological light-protective formulations containing triazine derivatives and one or several esters of unbranched-chain carboxylic acids and branched-chain alcohols**“

the specification of which is attached hereto.

-OR-

was filed on \_\_\_\_\_ as

Application Serial No. \_\_\_\_\_ and was amended \_\_\_\_\_

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations §1.56(a).

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)			Priority Claimed
<u>198 20 825.1</u>	<u>Germany</u>	<u>09. May 1998</u>	[X] yes [ ] no
(Number)	(Country)	(Day/Month/Yr. Filed)	
_____	_____	_____	[X] yes [ ] no
(Number)	(Country)	(Day/Month/Yr. Filed)	

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

<u>PCT/EP99/02709</u>	<u>22. April 1999</u>	<u>pending</u>
(Application Serial No.)	(Filing Date)	(Status)
		(patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punished by fine or imprisonment, or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

**POWER OF ATTORNEY:** As a named Inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

Kurt G. Briscoe, Reg. No. 33,141; William C. Gerstenzang, Reg. No. 27,552; Carmella A. O'Gorman, Reg. No. 33,749, and Stephen G. Ryan, Reg. No. 39,015 all of 660 White Plains Road, Tarrytown, New York 10591-5144; William R. Robinson, Reg. No. 27,224; Mark A. Montana, Reg. No. 44,948 all of 721 Route 202-206, Bridgewater, New Jersey 08807; Lorimer P. Brooks, Reg. No. 15,155; Davy E. Zoneraich, Reg. No. 37,267 all of 805 Third Avenue, 9<sup>th</sup> Floor, New York, NY 10022, my attorneys with full power of substitution and revocation.

**Send Correspondence To:**

Norris McLaughlin & Marcus, P.A.  
660 White Plains Road  
Tarrytown, N. Y. 10591-5144

**Direct Telephone Calls To:**

(914) 332-1700

Full Name Of Sole or First Inventor Heinrich GERS-BARLAG		Inventor's Signature <i>Heinrich Gers-Barlag</i>	Date 22/08/00
Residence Kiebitzgrund 11, 25495 Kummerfeld, Germany		Citizenship German	
Post Office Address Kiebitzgrund 11, 25495 Kummerfeld, Germany			
Full Name Of Second Inventor Rainer KRÖPKE		Inventor's Signature <i>Rainer Kröpke</i>	Date 28.08.00
Residence Achterndiek 23, 22869 Schenefeld, Germany		Citizenship German	
Post Office Address Achterndiek 23, 22869 Schenefeld, Germany			
Full Name Of Third Inventor		Inventor's Signature	Date
Residence		Citizenship	
Post Office Address			
Full Name Of Fourth Inventor		Inventor's Signature	Date
Residence		Citizenship	
Post Office Address			
Full Name Of Fifth Inventor		Inventor's Signature	Date
Residence		Citizenship	
Post Office Address			
Full Name Of Sixth Inventor		Inventor's Signature	Date
Residence		Citizenship	
Post Office Address			
Full Name Of Seventh Inventor		Inventor's Signature	Date
Residence		Citizenship	
Post Office Address			